On-Roadway In-Cabin Exposure to Particulate Matter: Measurement Results Using Both Continuous and Time-Integrated Sampling Approaches

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Abstract

The Atlanta Commuters Exposure (ACE) Study was designed to measure in-cabin exposure to roadway particulate pollution and acute health response in a panel of adults with and without asthma following a 2-h scripted route along major highways in Atlanta. This article focuses on methods and results of both continuous and integrated approaches used to measure the concentration of $PM_{2.5}$ mass, particle number concentration (PNC), black carbon (BC) mass, and particle-bound PAHs, in-cabin noise, PM elemental composition, elemental carbon, organic carbon, water-soluble organic carbon (WSOC) content, and speciation of a broad range of organic compounds including alkanes, hopanes, and PAHs. Speciated PM data indicates that in-cabin particles derive from three non-co-varying processes: the resuspension of road dust containing crustal elements and previously-deposited brake pad residue with a contribution of normal fuel combustion, incomplete combustion processes producing PAHs and carbon particles, and particles ablated from brake pads that have not previously deposited to the roadside environment. Most in-cabin pollutants were elevated during the warm season with the notable exception of PNC. PNC was not found to be correlated with most other pollutants. In-cabin concentrations were marginally higher when windows were open.

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1. INTRODUCTION

Exposure to traffic and traffic-related air pollution has been associated with adverse health, including respiratory responses such as decreased pulmonary function (Brunekreef et al. 1997; Gauderman et al. 2004; Sekine et al. 2004; McCreanor et al. 2007), increased respiratory symptoms (Bayer-Oglesby et al. 2006; Vigotti et al. 2007), and increased incidence of asthma or severity of asthma symptoms (Lwebuga-Mukasa et al. 2004; Zmirou et al. 2004; McConnell et al. 2006, 2010), as well as with cardiovascular outcomes such as increased levels of atherosclerosis (Hoffmann et al. 2007), alterations in heart rate variability (Riediker et al. 2004; Schwartz et al. 2005; Adar et al. 2007), and increased incidence of myocardial infarction (Peters et al. 2004; Lanki et al. 2006; Rosenlund et al. 2006; Tonne et al. 2007). Toxicological evidence suggests these associations are related to both the size and composition of traffic-related particulate matter (PM) (Brook 2008; Valavanidis et al. 2008; Møller et al. 2010). The number distribution of fresh vehicle emissions is dominated by particles in the ultrafine size range (<100 nm) (Kittelson 1998; Robert et al. 2007a,b), which have the highest deposition rates in the alveolar region of the lung (Heyder et al. 1986), and insoluble ultrafine particles are removed at a very slow rate (Stahlhofen et al. 1995; Möller et al. 2008). In addition, fresh traffic emissions contain constituents that are able to participate in oxidant-generating reactions in the airways, including transition metals, polycyclic aromatic hydrocarbons (PAHs), and other organic compounds (Chellam et al. 2005; Lough et al. 2005). The investigation of the mechanistic link between air pollution and health response is therefore facilitated by assessing exposure to oxidant-generating PM components and ultrafine PM (or its surrogate, particle number concentration, PNC).

Previous studies have investigated in-cabin exposures in a number of different settings. Rodes et al. (1998) measured the concentrations of PM mass, black carbon (BC), and a limited selection of metals and organics in vehicles on California roadways. Riediker et al. (2003, 2004) measured in-cabin $PM_{2.5}$, PAHs, elemental carbon and elements in North Carolina Highway Patrolmen along with cardiovascular response. Adar et al. (2007) measured $PM_{2.5}$ and BC exposure and cardiovascular response in a panel of senior citizens riding buses in St. Louis. Fruin et al. (2004) measured PNC, BC, and PAHs with a goal to identify vehicle, roadway, and driving characteristics that influence in-cabin concentrations. A review of studies examining transportation-related ultrafine particle exposures was published by Knibbs et al. (2011) and emphasized the need for further studies to elucidate the health effects of transit exposures. In light of these foundational studies, an urgent need continues to exist for detailed investigation of exposure to specific traffic-related pollutants etiologically-involved in the human health response.

The Atlanta Commuters Exposures (ACE) Study was established with a primary aim to investigate associations between in-cabin particulate pollution exposures and health responses in a panel of car commuters both with and without asthma. The ACE study utilized both continuous and integrated measures of PM, including methods reported by predecessor studies (e.g., PNC, BC, $PM_{2.5}$ mass, PM elemental composition) as well as methods not previously applied to in-cabin studies (e.g., speciation of organic compounds including alkanes, hopanes, and PAHs from integrated filter samples). In addition, in-cabin noise level was continuously monitored, and a corresponding suite of respiratory and

cardiovascular endpoints was measured at multiple time points following each commute. The ACE study included an initial study funded by the Centers for Disease Control and Prevention (referred to as ACE-1) and a follow-up study funded by the Environmental Protection Agency (ACE-2). The current manuscript presents data from ACE-1 with a focus on the magnitude, composition, and nature of the measured in-cabin exposures. Human health response measured during the commutes and associations between the health and pollutant measurements will be presented in companion manuscripts.

2. METHODS

2.1. Study Overview

In-cabin concentrations of PM and PM components were measured for 42 subjects, each of whom were to complete two commutes during the protocol for a total of 84 scheduled commuting sessions. Commutes were conducted in each subject's personal vehicle and followed a scripted route of approximately 2 h duration during the morning rush period in Atlanta (~7–9 am). Commute routes were selected to maximize the time spent on controlled-access freeways (Figure S1 in the online supplementary information [SI]) and were conducted during all seasons (2010–2011) and in all meteorological conditions. Vehicle windows were alternately opened and closed every 15 min throughout the commute. Compliance was voluntary and was greater when the weather was pleasant. While windows were closed, subjects were permitted to use the vehicle's air conditioner or ventilation system set to outside air rather than the recirculation setting. Additional details describing the study design and implementation may be found in the SI.

2.2. Continuous Measurements

The continuous instruments used for this study are listed in Table 1. These devices were placed in a sampling tray located in the front passenger seat of the vehicle with sample inlets directed toward the middle of the vehicle cabin and no more than 1 m from the breathing zone of the driver (Figure S2). In addition, vehicle location and speed was continuously recorded with a GPS device, and a small camera was affixed to the inside of the windshield facing the direction of travel and recorded a still image of traffic conditions every 2 s. A more detailed description of continuous instrumentation is available in the SI.

2.3. Integrated Measurements

Integrated measurements of particle mass and composition were collected using filter holders and a cascade impactor installed in the front passenger seat. Flow through these filters was provided by vacuum pumps installed in the trunk of the vehicle and powered by two deep-cycle lead-acid batteries also installed in the trunk. Exhaust from the pumps was filtered and diverted to the exterior of the vehicle. Flow was controlled using critical orifices, and flowrate measurements were performed at the beginning and end of each commute. Additional details are included in the SI.

2.3.1. PM Speciation—Fine mode PM speciation analysis included measurements of elemental composition, organic speciation (n-alkanes, hopanes, and PAHs), organic and elemental carbon (OC and EC) mass concentration and water-soluble organic carbon

(WSOC). The enrichment factor (EF) for elements was calculated as the ratio of an element to aluminum in the filter sample divided by the ratio of that same element to aluminum in a representative soil sample from Georgia. Additional details are available in the SI.

2.4. Data Analysis

Descriptive statistics and correlation analyses for each pollutant measure were calculated for all commutes and then for commutes stratified by warm season (15 April–14 October) or cold season (15 October–14 April). Similar analyses were conducted for each continuous pollutant parameter stratified by window open/closed status. Factor analysis of integrated particle species was conducted for dimension-reduction purposes using a varimax rotation method to organize all PM speciated constituents into a small number of groups of co-varying groups. Statistical analyses were conducted using SAS v9.2 (SAS Institute Inc., Cary, NC, USA).

3. RESULTS AND DISCUSSION

3.1. Study Overview

A total of 81 commutes were completed with 41 (51%) occurring during the cold season and 40 (49%) during the warm season. Data completeness, defined as the ratio of samples successfully collected to the number of expected samples, is listed in Table 1. Additional details are included in the SI.

3.2. Integrated Measurements

A summary of filter-based measurements is presented in Table 2, including a selection of results from speciation analysis. A total of 19 field blanks were collected and analyzed for each of the filter-based sample lines (i.e., 23% of total filters collected), and the method limit of detection (LOD) for each parameter was defined as three times the standard deviation of the field blanks. For many pollutant parameters, field blank values were below the instrumental detection limit, and the method LOD for these parameters was assumed to be the instrumental LOD. The current results have been blank-corrected.

3.2.1. EC-OC Analysis—The assessment of semi-volatile OC was not an original primary aim of ACE-1, and sample lines were therefore not denuded. OC concentrations measured using the TOT method consequently comprised a very large proportion or even exceeded the gravimetrically-derived $PM_{2.5}$ concentrations (18–650%, median = 110%). This positive artifact is due to the adsorption of semi-volatile OC compounds onto the high surface area of the quartz fiber substrate of the OC filters (Mader and Pankow 2001). Previous studies have found the positive OC artifact to be 10–60% of the OC concentration (Turpin et al. 2000; Subramanian et al. 2004), and to be higher when sampling fresh, less photo-chemically oxidized emissions (Baumann et al. 2003) or when sampling for short durations with little time for the filter to reach gas-particle phase equilibrium (Turpin et al. 2000). Both of these conditions were present during the ACE-1 sampling sessions and likely contribute to the elevated OC artifacts observed.

Mean in-cabin EC mass measured during the commutes was 2.8 ± 1.9 μg·m^{-3}, and OC mass was 19.2 ± 6.9 μg·m^{-3}. These concentrations similar to or exceed previously reported non-denuded roadside measurements. Weber et al. (2007) reported the EC concentration as 3.6 ± 1.1 μg·m^{-3} and OC as 10.8 ± 1.6 μg·m^{-3} in summer alongside I-75/85 in Atlanta (Weber et al. 2007), and Yan et al. (2009) report summertime EC as 4.1 ± 1.1 μg·m^{-3} and OC as 8.2 ± 1.2 μg·m^{-3} and wintertime EC as 2.7 ± 1.3 μg·m^{-3} and OC as 5.1 ± 2.5 μg·m^{-3} at the same location (Yan et al. 2009). This location is situated at the downtown portion of the route typically used by the current study. In comparison to other cities, our in-cabin measurements are comparable yet slightly higher than nondenuded measurements along similar style roadways in Los Angeles (EC = 1.8 ± 1.2 μg·m^{-3}, OC = 14.9 ± 5.2 μg·m^{-3}) (Phuleria et al. 2007) and Hong kong (OC = 13.4 ± 7.9 μg·m^{-3}) (Cheng et al. 2010). It should be noted that the referenced values reflect 24 h integrated averages (Yan et al. are 12 h averages) whereas our measurements are 2 h averages conducted at the time of peak traffic volume. The mean ratio of EC/OC for in-cabin measurements was 0.14 ± 0.08, whereas it was 0.33 and 0.50 as measured in Atlanta by Weber et al. (2007) and Yan et al. (2009) respectively. Plausible explanations for our lower observed EC/OC ratios are that (a) our in-cabin samples were more influenced by a positive OC artifact than the referenced 12- or 24-h roadside measurements, (b) the fraction of vehicles that are diesel is lower during the morning rush hour than the 24-h average (as noted in Georgia Department of Transportation data), and (c) sampling for this study was conducted after implementation had begun for the EPA 2007 Heavy-Duty Highway Rule, which requires new heavy-duty diesel trucks to be fitted with a diesel particulate filter. Vehicles complying with the 2007 rule have much lower EC emissions than vehicles meeting the previous standard (Khalek et al. 2011).

3.2.2. WSOC Analysis—The mean in-cabin WSOC concentration was observed to be 6.2 ± 3.9 μg·m^{-3}, values that are comparable to other non-denuded roadside measurements. Weber et al. (2007) reported 4.7 ± 0.7 μg·m^{-3} of WSOC along I-75/85 in Atlanta (Weber et al. 2007), Ho et al. (2006) measured $2.1 \cdot 0.3$ μg·m^{-3} during summer and 3.5 ± 0.5 μg·m^{-3} during winter at a roadside location in Hong Kong (Ho et al. 2006), and Bao et al. (2009) measured 6.2 μg·m^{-3} in summer and 2.5 ± 0.5 μg·m^{-3} in in winter at a roadside location Japan (Bao et al. 2009). Our in-cabin measurements are higher than WSOC concentrations reported for urban settings that are more distant to large roadways. Nonroadside measurements in Atlanta range between 0.5–3.0 μg·m^{-3} and vary depending on time of year and distance from ·road (Hennigan et al. 2009; Zhang et al. 2012a,b). The WSOC/OC ratio for the ACE-1 study was 0.36 ± 0.28, a value that is similar to what is typically observed in other ambient measurement settings. Previous measurements in the Atlanta area suggest a WSOC/OC ratio typically occurring in the 0.4–0.6 range (Weber et al. 2007; Zhang et al. 2012b). Ongoing speciation of the carbon fraction on the filters, including a characterization of the brown carbon content, may provide additional insight into the source of in-cabin WSOC measured during these commutes.

3.2.3. Factor Analysis of Elements and Organic Species—In-cabin concentrations of most PM elements were comparable to and within a factor of 2 of the in-cabin measurements reported by Riediker et al. (2003) that used X-ray fluorescence to quantify elemental concentrations. An earlier study by Rodes et al. (1998) found in-cabin

samples for most elements to be below LOD, although the few overlapping species that were within their detectable range are within a factor of 5 of the current results. In addition, our in-cabin crustal element concentrations are similar to those previously reported in other roadside microenvironments (Ntziachristos et al. 2007; Hays et al. 2011), while the concentrations of anthropogenic elemental species (EF > 100) were a factor of 2–3 higher than measurements from a roadside location in North Carolina (Hays et al. 2011). To our knowledge, these are the first reported measurements of in-cabin concentrations of particulate organic species during typical commuting activities. In-cabin concentrations of hopanes, alkanes, and PAHs were similar to long-term averages at the St. Louis supersite (Sheesley et al. 2007), but were substantially higher than roadside measurements in North Carolina (Olson and McDow 2009). The predominant in-cabin PAH species reported here are fluoranthene, pyrene, benzo[b+k]fluoranthene, and benzo[a]pyrene. This profile is similar to the St. Louis and North Carolina studies, although fluoranthene was not reported in North Carolina and the St. Louis study reported relatively high levels of indeno[1,2,3-cd]pyrene, which was below LOD in 67% of the current samples.

Initial factor analysis was conducted separately for each class of speciated components (e.g., elements, PAHs, alkanes), and it was observed that among the speciated PAHs, fluoranthene and pyrene were highly correlated with one another and comprised a single PAH factor while benz[a]anthracene, chrysene, benzo[b+k]fluoranthene, benzo[a]pyrene, benzo[e]pyrene, and coronene comprised a another PAH factor. These groupings are consistent with previous findings using principal component or factor analysis (Guo et al. 2003; Ravindra et al. 2008) and loosely correspond to factors related to diesel and gasoline combustion respectively. Fluoranthene and pyrene have a lower molecular weight and higher volatility than the other measured PAHs, raising the possibility that these groupings may be temperature-dependent. Given that these samples were collected in-cabin, we did not observe a significant difference in cabin temperature or fluoranthene concentration between seasons.

For the elemental species, crustal elements comprised a primary factor, with copper, zinc, and lead comprising a secondary factor. For the speciated organic compounds, it was found that the sum of speciated hopanes comprised the primary factor and the sum of n-alkanes with 23–27 carbons comprised a secondary factor. Based on these initial analysis results, factor analysis for all classes of compounds was performed and included input variables as illustrated in Figure 1. This selection of variables captured most of the variance in the dataset with the least amount of missing data ($N = 50$ for factor analysis). Final factor analysis did not include variables of which other measured parameters comprised a subset (e.g., EC, OC, WSOC, and $PM_{2.5}$ mass); however, exploratory factor analysis modeling was performed including one or more of these variables.

The largest factor (explaining 47% of the variance) was dominated by crustal elements and other species that are typically present in resuspended road dust. The conventional EF threshold value for crustal elements is 10 (Duce et al. 1975) though elements with EF = 10–100 also have a strong crustal component. By this metric, the dominant crustal elements were Fe (EF = 19.8), Al (EF = 1.0 by definition), and Ca (EF = 3.9). Other crustal elements with modest concentrations also exhibited factor loadings of approximately 0.7 or greater

with factor 1, including K, Mg, Mn, and Ti. During exploratory model runs including OC as a variable, OC was most closely associated with factor 1 (factor loading = 0.70).

Notably, S, Ba, and Sb were also shown to have large loadings with factor 1 and were strongly correlated with crustal elements. These elements are highly enriched with EFs several orders of magnitude greater than 1.0 and are present in brake pad material (Schauer et al. 2006; Thorpe and Harrison 2008), while S is also present in tire material, lubricating oil and sulfate aerosol. Given that the variance for S, Ba, and Sb closely matches that of the main crustal species, we believe that particles containing these elements had previously deposited in the roadside environment and that the primary source of Ba and Sb in the in-cabin microenvironment is the resuspension of road dust. The factor loading for S is somewhat lower than for the other species in factor 1, and correlations between S and the main crustal elements are similarly less than between Ba or Sb and crystals. This suggests that though resuspended dust is likely to be an important source of S, other sources such as sulfate aerosol or lubricating oil may be contributing to S concentrations. It is interesting to note that Ba and Sb have a variance structure matching that of crustal elements rather than that of other elements with a brake pad source such as Cu, Zn, and Pb. A previous analysis of the difference in composition between brake pad bulk material and generated dust suggested that some brake pad components are emitted directly into the air while others form a "transfer film" at the disc/pad interface that subsequently dislodges to the roadway (Thorpe and Harrison 2008). This explanation is consistent with an interpretation that in the current study, Ba and Sb deposit to road dust before being resuspended whereas a higher proportion of Cu, Zn, and Pb enter the vehicle cabin directly following the braking action of nearby vehicles, including the one being driven.

The next largest factor (explaining 18% of the variance) was dominated by PAHs. Initial factor analysis including just PAH species indicated that fluoranthene and pyrene comprised a separate factor from other PAHs. This finding is reflected in the modest factor loading on factor 2 for both fluoranthene (0.58) and pyrene (0.23). Despite this, these PAHs were included in factor 2 given that their factor loading is near zero or negative for all other factors. During exploratory model runs including EC as a variable, EC was strongly associated with this factor (factor loading = 0.82). In addition, it was found that although hopanes and alkanes were not strongly associated with any factor, the highest factor loading for hopanes was with factor 1 while that of alkanes was with factor 2. Although these are both markers of petroleum combustion (Schauer et al. 1996), our findings suggest that for ACE-1, factor 1 included a contribution of normal fuel combustion (reflected by hopanes and OC factor loadings) while factor 2 reflected a contribution of incomplete fuel combustion (underscored by PAHs and EC factor loadings). Furthermore, a third factor (explaining 12% of the variance) was comprised of copper and zinc with a smaller contribution from lead, indicative of brake pad and tire wear (Thorpe and Harrison 2008).

The results from the current factor analysis, which were novel in utilizing a broad suite of elemental and organic particulate components, suggest that for the ACE-1 commutes, in-cabin $PM_{2.5}$ was impacted by three processes that do not necessarily co-vary: resuspension of road dust with a contribution of normal fuel combustion, incomplete combustion of petroleum fuels, and mechanical generation of particles from brake pad and tire wear. All

three processes are influenced by meteorological and traffic conditions. Factor 1 would conceptually be maximized by dry conditions with free-flowing traffic, factor 2 is consistent with slow or stopped traffic and a larger proportion of diesel vehicles, and factor 3 is consistent with stop-and-go traffic with disproportionate braking action.

3.3. Continuous Measurements

Continuous measurements of BC, pbPAHs, PNC, and $PM_{2.5}$ mass were conducted with a time resolution of 1 min or less. The mean values of these measurements are similar to those previously reported for in-cabin (Riediker et al. 2003; Adar et al. 2007; Zuurbier et al. 2010) or roadside (McCreanor et al. 2007) air pollution exposures. Notably, the primary method for measuring PNC in this study (TSI Inc. P-Trak) had a lower cutpoint of 20 nm whereas many of the referenced studies used an instrument with a 10-nm cutpoint. For 31 of the 84 commutes in our study, both instruments were used, and in this subset, $29 \pm 15\%$ of PNC was observed to be in the 10–20 nm size range. The mean concentration of pbPAH for the current study was a factor of 5 higher than that reported by Riediker et al (2003), though that study was conducted in a less populated area, at different times of day, with traffic volumes that were likely lower than those in the current study (Table 3). Our in-cabin values were very similar to those measured on the I-405 freeway in Los Angeles and were a factor of 2 lower than those on the more diesel-influenced I-710 freeway (Westerdahl et al. 2005; Zhu et al. 2008). These values were also comparable to morning rush hour measurements made at a nonroadside location in Mexico City (Marr et al. 2006; Thornhill et al. 2008).

3.3.1. Relationship Between Continuous and Integrated Measurements—The ACE-1 sampling design included an integrated measurement that closely corresponded to the BC, pbPAHs, and $PM_{2.5}$ mass continuous measurements. We conducted linear regression analyses for these measurements by pairing the average of the continuous measure for a commute with the integrated measure of the same commute. For $PM_{2.5}$ mass, the proxy continuous measure based on fine mode number concentration is by definition correlated with integrated $PM_{2.5}$ mass since it is scaled by a density estimate derived from the integrated measure; however, the correlation of continuously-measured volume to integrated mass concentration was modest (Pearson's $r = 0.56$). For BC, the continuous measurements were integrated by comparing the attenuation across the instrument's internal filter at the beginning and end of the commute rather than averaging the 1-min values. There was modest correlation between this integrated value and filter-based EC (Pearson's $r = 0.72$) (Figure S3) (in the online SI). The slope of the linear regression model was close to 1 (slope 1.1, $p[H_0:\text{slope } 1] = 0.37$) though the intercept was 2.8 ± 0.41 $\mu\text{g}\cdot\text{m}^{-3}$. Given that the TD-GC/MS analysis of filter samples and the continuous measurement of pbPAHs employ very different methodologies, a comparison of these approaches must be interpreted cautiously. A discussion of this comparison is included in the SI.

3.3.2. Correlation of Continuous Measurements with Each Other—Correlation analysis was performed for each continuous measure with the other continuous parameters (Table 4). For the 1-min data, the large number of data points leads to all correlation coefficients being different than zero with $p < 0.0001$; however, the associations among most pollutants were modest to weak on this time scale. In particular, in-cabin $PM_{2.5}$ was weakly

associated with the other continuously measured pollutants ($r^2 = 0.03$–0.06), whereas PNC exhibited a slightly stronger association with BC and pbPAH ($r^2 = 0.12$–0.14). This was largely expected since both BC and pbPAH share a common source and were associated with each other ($r^2 = 0.56$). The correlations of commute averages were stronger than the 1-min averages for $PM_{2.5}$ and all other pollutants but were nearly the same for PNC with BC and pbPAH. This change in correlation coefficient with time scale is in general agreement with that observed in a St. Louis bus study (Adar et al. 2007).

Descriptive statistics for in-cabin concentrations stratified by window status (open or closed) are shown in Table 5. Although this study was not designed to quantify the influence of specific exposure factors on in-cabin pollutant concentration as done in other studies (Fruin et al. 2008; Hudda et al. 2011), we did collect sufficient data to examine differences in in-cabin concentrations by window status. A 50 commutes complied with the window-opening protocol, though it should be noted that the window-open periods were not representative since study subjects were more likely to comply when temperatures were mild and there was no precipitation (54% of cool season commutes were compliant compared to 61% of the warm season commutes). The difference in median concentration for in-cabin $PM_{2.5}$ was 28% higher when windows were open; for PNC, the increase was 24%; for BC, it was 18%, and for pbPAH, it was 7%. In addition, although PM_{coarse} (D_p 2.5–10 μm) mass was not measured for this study, the largest diameter channel on the AeroTrak roughly corresponds to this size range, and the difference in the number concentration measured by this channel was 26% higher during open window periods. These results are consistent with previous findings that vehicle air filters can modestly reduce in-cabin particle concentrations, though there is much variation with vehicle model and age and particles in the 0.1–1.0 μm size range have less than 50% filtration efficiency (Qi et al. 2008). Figure 2 shows the time series for $PM_{2.5}$ and PNC for the vehicle with the most pronounced difference in concentration between windows open and closed periods. For this vehicle, the in-cabin $PM_{2.5}$ concentration when the windows were open is 110% higher than when the windows are closed. For PNC, although values were 44% higher when windows were open, there were still time periods with high PNC when the windows were closed. This is consistent with previous studies which have found that ultrafine particles efficiently infiltrate car cabins even when windows are closed and circulation air is filtered (Fruin et al. 2008; Hudda et al. 2011).

3.4. Seasonal Variation

Several important pollutant parameters, both integrated and continuous, exhibited seasonal variation in concentration as well as in their correlation patterns with other pollutants. For the purposes of this analysis, we defined the warm season as 15 April–14 October and the cool season as 15 October–14 April. A summary of the seasonal variability is presented in Table 6.

Most of the measured pollutant values were higher during the warm season, though this was only statistically significant for continuously measured pollutants. The exceptions were OC, WSOC, and PNC, which were higher in the cool season, with only PNC exhibiting statistically significant seasonal differences. Some of these seasonal differences are likely

influenced by window status given that window-opening compliance was greater during the warm season. During the warm season, $PM_{2.5}$ and Fe (representing crustal elements) were highly correlated with WSOC ($r = 0.83$ and 0.76 respectively, $p < 0.0001$ for both); however, this correlation is not present in the cool season. The magnitude of the correlation and the strength of the p-value suggest this is not a chance finding. The only other warm season correlated pollutants share a source and are chemically similar (e.g., EC, BC, and pbPAH). During the cool season, most parameters were reasonably well-correlated with each other with significant or marginally-significant p-values. The exception in this case is WSOC, which was weakly or negatively-correlated with all other pollutants. Both of these seasonal trends reflect the importance of biogenic sources of OC and WSOC in this region. In the cool season, especially during the morning rush period, the mixing height is low, inducing greater covariance among most anthropogenic pollutants due to the effect of a reduced mixing volume. Although cool season biomass burning contributes somewhat to WSOC concentration, WSOC was not shown to co-vary in the same manner as other pollutants in the absence of biogenic sources.

4. SUMMARY AND CONCLUSIONS

The ACE-1 Study measured the in-cabin concentration of a wide-range of specific pollutants in a variety of cars operating under typical driving conditions for Atlanta highways. This was the first study to include measurements of elemental and organic speciation of PM, elemental and organic carbon, WSOC, and time-resolved $PM_{2.5}$, PNC, BC, and pbPAH in a study of this size. Factor analysis of speciated data indicates the influence of three non-co-varying processes: resuspension of road dust with a contribution of normal fuel combustion, incomplete combustion of petroleum fuels, and the infiltration of nondeposited brake pad particles into the vehicle cabin. PNC showed very modest correlation with other pollutant measures and was unique in being significantly-higher in the cool season. Further analysis of this exceptionally large data set is ongoing, and companion manuscripts will present results of health outcome measures and detailed examination of exposure data.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.
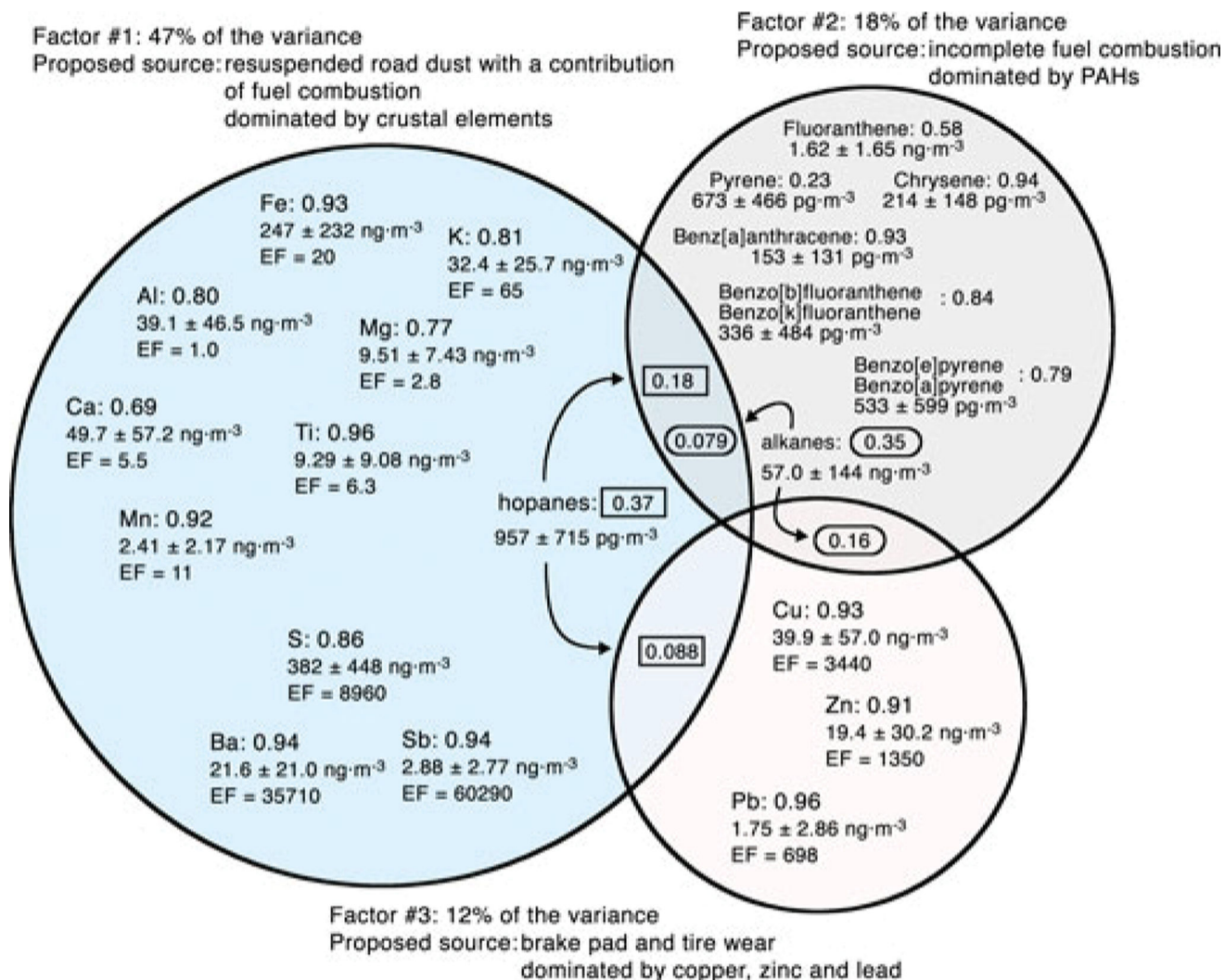
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Factor #1: 47% of the variance
Proposed source: resuspended road dust with a contribution of fuel combustion dominated by crustal elements

Factor #2: 18% of the variance
Proposed source: incomplete fuel combustion dominated by PAHs

Fe: 0.93
247 ± 232 ng·m⁻³
EF = 20

K: 0.81
32.4 ± 25.7 ng·m⁻³
EF = 65

Al: 0.80
39.1 ± 46.5 ng·m⁻³
EF = 1.0

Mg: 0.77
9.51 ± 7.43 ng·m⁻³
EF = 2.8

Ca: 0.69
49.7 ± 57.2 ng·m⁻³
EF = 5.5

Ti: 0.96
9.29 ± 9.08 ng·m⁻³
EF = 6.3

Mn: 0.92
2.41 ± 2.17 ng·m⁻³
EF = 11

Fluoranthene: 0.58
1.62 ± 1.65 ng·m⁻³

Pyrene: 0.23
673 ± 466 pg·m⁻³

Chrysene: 0.94
214 ± 148 pg·m⁻³

Benz[a]anthracene: 0.93
153 ± 131 pg·m⁻³

Benzo[b]fluoranthene
Benzo[k]fluoranthene : 0.84
336 ± 484 pg·m⁻³

Benzo[e]pyrene
Benzo[a]pyrene : 0.79
533 ± 599 pg·m⁻³

0.18

0.079   alkanes: 0.35
57.0 ± 144 ng·m⁻³

hopanes: 0.37
957 ± 715 pg·m⁻³

0.16

S: 0.86
382 ± 448 ng·m⁻³
EF = 8960

0.088

Cu: 0.93
39.9 ± 57.0 ng·m⁻³
EF = 3440

Zn: 0.91
19.4 ± 30.2 ng·m⁻³
EF = 1350

Ba: 0.94
21.6 ± 21.0 ng·m⁻³
EF = 35710

Sb: 0.94
2.88 ± 2.77 ng·m⁻³
EF = 60290

Pb: 0.96
1.75 ± 2.86 ng·m⁻³
EF = 698

Factor #3: 12% of the variance
Proposed source: brake pad and tire wear dominated by copper, zinc and lead

FIG. 1.
Factor analysis of integrated samples (N = 50). PM constituents are labeled by name and factor loading followed by the mean ± standard deviation concentration and enrichment factor (for elements). Hopanes and alkanes are not unambiguously associated with any factor, and factor loadings are shown for all three factors. Hopanes are the sum of 17α(H),21β(H)-29-norhopane and 17α(H),21β(H)-hopane. Alkanes are the sum of C23-C27 n-alkanes.
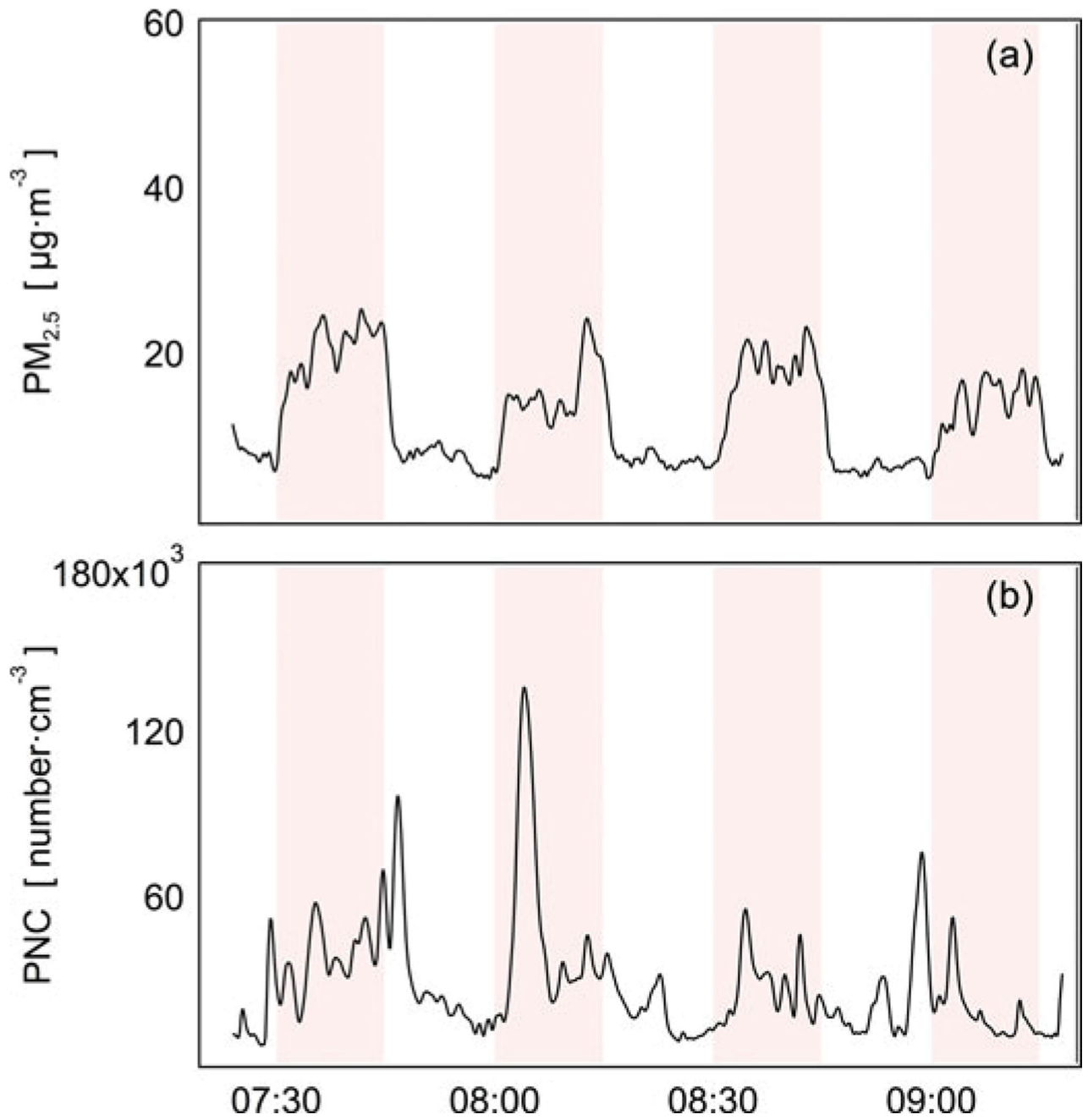
FIG. 2.
Time series analysis for PM2.5 (a) and PNC (b) for an atypical commute with discernible cycles between windows open and closed. Shaded areas are when windows are open.

TABLE 1

In-cabin exposure parameters measured during the ACE-1 study: both continuous and time-integrated approaches

	Parameter	Method	Time resolution	Flow rate [L·min^{-1}]	Instrumentation	Completeness
Continuous measures	Black carbon mass concentration	Filter transmittance	1 min	0.1	MicroAethAE51 (AethLabs, San Francisco, CA)	0.93
	Particle-bound PAH mass concentration	Photoionization	10 s	1.0	PAS 2000CE (EcoChem Analytics, League City, TX)	0.93
	Particle number concentration (D_p > 20 nm)	Condensation particle counter	1 s	0.1	P-Trak (TSI Inc., Shoreview, MN)	0.91
	Particle number concentration (D_p > 10 nm)	Condensation particle counter	1 s	0.1	CPC model 3007 (TSI Inc., Shoreview, MN)	0.38
	Calculated PM$_{2.5}$ mass concentration	Optical particle counter	1 s	2.83	AeroTrak model 9306 (TSI Inc., Shoreview, MN)	0.92
	Noise	Sound level meter	1 s	NA	Extech HD600 (Extech Instruments, Nashua, NH)	0.82
PM$_{2.5}$ Integrated Measures	Fine mode mass	Gravimetric		30	XS3DU balance (Mettler-Toledo, Columbus, OH)	0.86
	Elemental composition	ICP-MS[a]			PQ Excell (ThermoElemental, Waltham, MA)	0.91
	Organic speciation	TD-GC/MS[b]		30	GC-MS model 5973 (Agilent Tech., Santa Clara, CA)	0.91
	Elemental and organic carbon	TOT[c]		15	Lab OC-EC (Sunset Labs, Hillsborough, NC)	0.89
	Water-soluble organic carbon	TOC[d]			Sievers 900 Portable (GE Analytical, Boulder, CO)	0.87

[a] Inductively-coupled plasma-mass spectrometry, University of Wisconsin.

[b] Thermal desorption/gas chromatography-mass spectrometry, University of Wisconsin.

[c] Thermal-optical transmittance, University of Wisconsin and Georgia Tech.

[d] Total organic carbon analyzer, Georgia Tech.

TABLE 2

Descriptive statistics of selected integrated filter-based measurements. Statistics only reflect values above LOD

	Parameter	Unit	Mean (SD)	Median (IQR)	N	Percent above LOD	Enrichment factor
	PM$_{2.5}$	μg·m^{-3}	19.2 (13.6)	15.2 (14.2)	72	89	-
	EC	μg·m^{-3}	2.8 (1.9)	2.3 (2.4)	76	94	-
	OC	μg·m^{-3}	19.2 (6.9)	18.6 (8.9)	75	93	-
	WSOC	μg·m^{-3}	6.2 (3.9)	5.6 (3.2)	73	90	-
PAHs	benzo[a]pyrene	pg·m^{-3}	333 (434)	225 (296)	61	75	-
	benzo[a]pyrene	pg·m^{-3}	333 (434)	225 (296)	61	75	-
	pyrene	pg·m^{-3}	673 (466)	542 (498)	76	93	-
	benzo[b+k]fluoranthene	pg·m^{-3}	336 (484)	191 (285)	56	69	-
	benzo[ghi]perylene	pg·m^{-3}	231 (297)	126 (206)	51	63	-
	fluoranthene	pg·m^{-3}	1615 (1646)	1053 (1169)	76	94	-
	benz [a] anthracene	pg·m^{-3}	153 (131)	121 (148)	71	88	-
	chrysene	pg·m^{-3}	214 (148)	174 (187)	75	93	-
	Sum of speciated hopanes[a]	pg·m^{-3}	956 (715)	733 (769)	73	90	-
	Sum of C23-C27 alkanes	ng·m^{-3}	57.0 (144)	34.8 (25.7)	75	93	-
Elements	Sodium	ng·m^{-3}	32.3 (29.3)	22.8 (30.2)	47	58	7.0
	Magnesium	ng·m^{-3}	9.51 (7.43)	8.45 (8.09)	72	89	2.8
	Aluminum	ng·m^{-3}	39.1 (46.5)	23.5 (28.1)	67	83	1.0
	Sulfur	ng·m^{-3}	382 (448)	253 (272)	74	91	9.0×10^3
	Potassium	ng·m^{-3}	32.4 (25.7)	27.4 (32.2)	73	90	65
	Calcium	ng·m^{-3}	49.7 (57.2)	33.0 (39.8)	59	73	5.5
	Titanium	ng·m^{-3}	9.29 (9.08)	7.14 (9.89)	66	81	6.3
	Manganese	ng·m^{-3}	2.41 (2.17)	1.72 (2.21)	73	90	11
	Iron	ng·m^{-3}	247 (232)	193 (265)	74	91	20
	Copper	ng·m^{-3}	39.9 (57.0)	20.7 (26.4)	68	84	3.4×10^3
	Zinc	ng·m^{-3}	19.4 (30.2)	8.19 (15.8)	67	83	1.3×10^3
	Antimony	ng·m^{-3}	2.88 (2.77)	2.21 (2.75)	65	80	6.0×10^4
	Barium	ng·m^{-3}	21.6 (21.0)	17.5 (18.9)	74	91	3.6×10^4

Parameter	Unit	Mean (SD)	Median (IQR)	N	Percent above LOD	Enrichment factor
Lead	ng·m^{-3}	1.75 (2.86)	0.807 (1.22)	71	88	698

[a]Sum of 17α(H),21β(H)-29-norhopane and 17α(H),21β(H)-hopane.

TABLE 3

Descriptive statistics for continuous measurements and comparison to similar exposure studies

Parameter	Unit	Mean (SD)	NC patrolmen study[b]	St. Louis bus study[c]	Dutch Commuters study[d]	Oxford Street study[e]
$PM_{2.5}$	$\mu g \cdot m^{-3}$	25.1 (13.4)	24.1 (14.6)	17.2 (10.3)	115 (118)	28.3
PNC	$\# \cdot cm^{-3}$	2.6×10^4 (1.2×10^4)	Not measured	Not measured	4.1×10^4 (1.0×10^4)	6.4×10^4
BC or EC	$\mu g \cdot m^{-3}$	6.6 (3.2)	2.3 (0.8)	2.9 (2.5)	14.7 (4.6)	7.5
pbPAH	$ng \cdot m^{-3}$	119 (32.3)	21.5 (10.3)	Not measured	Not measured	Not measured

[a]Mean and SD across commutes of the within commute maximum value.

[b]Riediker et al. (2003).

[c]Adar et al. (2007), showing results for the bus portion of the study.

[d]Zuurbier et al. (2010), showing results for the gasoline car portion of the study, measured PNC using a TSI Model 3007 (minimum D_p = 10 nm), measured filter absorbance and converted to EC according to Cyrys et al. (2003).

[e]McCreanor et al. (2007), showing median values for the Oxford Street portion of the study, measured PNC using a TSI Model 3007 (minimum D_p = 10 nm).

TABLE 4

Correlation of continuous data[a]

	PM2.5	**PNC**	**BC**	**pbPAH**
PM$_{2.5}$		0.18	0.25	0.20
		7314	7350	7467
		<0.0001	<0.0001	<0.0001
PNC	0.34		0.37	0.35
	73		8093	7990
	0.0032		<0.0001	<0.0001
BC	0.50	0.26		0.75
	75	74		8008
	<0.0001	0.0256		<0.0001
pbPAH	0.43	0.31	0.86	
	75	74	76	
	<0.0001	0.0067	<0.001	

[a]First row is Pearson's correlation coefficient; second row is N; third row is p-value for the correlation. Above the blank boxes are the correlations of 1-min data, and below the blank boxes are correlations of commute averages.

TABLE 5

Influence of window status (open or closed) on in-vehicle concentrations

	Closed		Open		
	Mean (SD)	**Median (IQR)**	**Mean (SD)**	**Median (IQR)**	**p-value**
PM$_{2.5}$	17.8 (13.1)	14.6 (12.4)	20.9 (13.1)	18.8 (15.8)	<0.0001
PNC	27,068 (23,534)	20,503 (23,805)	31,373 (26,406)	25,485 (24,781)	<0.0001
BC	6.1 (6.3)	4.5 (5.6)	7.5 (8.1)	5.3 (7.2)	<0.0001
pbPAH	116 (83.5)	99.5 (82.4)	127 (97.8)	107 (84.5)	<0.0001

TABLE 6

Correlation matrix of seasonal variation of in-vehicle pollutants[a].

Cool→ Warn↓	PM$_{2.5}$ mass 13.1(12.5)[b]	EC 1.94(2.28)[b]	OC 19.9(10.3)[b]	Wsoc 7.33(4.64)[b]	Fe 168(216)[c]	Cu 22.6(32.0)[c]	BC 4.81(3.85)[b]	PNC 30.4(15.7)[d]	pbPAH 102(41.2)[c]
PM$_{2.5}$ mass 16.9(14.1)[b]	33% 0.093 70	0.74 <0.0001 36	0.51 0.0014 36	0.23 0.19 35	0.52 0.0010 36	0.36 0.032 35	0.57 0.0003 36	0.66 <0.0001 34	0.73 <0.0001 36
EC 2.68(3.02)[b]	0.014 0.94 35	36% 0.056 74	0.58 0.0001 39	0.070 0.67 39	0.67 <0.0001 39	0.43 0.0081 37	0.68 <0.0001 40	0.60 <0.0001 38	0.89 <0.0001 40
OC 17.2(7.09)[b]	0.0093 0.96 35	0.59 0.0002 36	−8.6% 0.28 73	0.031 0.85 38	0.56 0.0002 38	0.59 0.0001 37	0.30 0.11 39	0.27 0.10 37	0.40 0.012 39
WSOC 4.71(2.45)[b]	0.83 <0.0001 33	−0.17 0.35 33	−0.085 0.64 33	−4.4% 0.88 71	0.034 0.84 38	0.18 0.49 36	−0.0033 0.98 39	0.0076 0.96 37	−0.0047 0.98 39
Fe 203(307)[c]	0.78 <0.0001 35	0.17 0.33 35	0.036 0.84 35	0.76 <0.0001 33	53% 0.51 73	0.50 0.0018 37	0.52 0.0006 39	0.32 0.057 37	0.61 <0.0001 39
Cu 20.3(24.3)[c]	0.34 0.043 35	−0.18 0.50 35	0.38 0.025 35	0.29 0.098 33	0.29 0.092 35	7.2% 0.84 71	0.38 0.020 37	0.27 0.12 35	0.36 0.028 37
BC 7.44(5.43)[b]	0.032 0.86 33	0.61 0.0002 33	0.32 0.068 33	−0.18 0.33 31	0.14 0.43 33	−0.10 0.56 33	49% 0.0002 76	0.64 <0.0001 35	0.82 <0.0001 37
PNC 21.3(11.8)[d]	0.060 0.74 33	0.010 0.96 33	0.24 0.18 33	−0.022 0.91 31	0.14 0.44 33	0.39 0.024 33	0.37 0.12 35	−27% 0.0029 74	0.70 <0.0001 38
pbPAH 129(47.1)[c]	−0.052 0.77 34	0.58 0.0003 34	0.32 0.069 34	−0.22 0.22 32	0.034 0.85 34	−0.095 0.59 34	0.86 <0.0001 36	0.17 0.32 36	20% 0.0030 76

[a] Values above the diagonal (italicized) are cool season (15 October–14 April), values below the diagonal (italicized) are warm season (15 April–14 October). For both, the top line is Pearson correlation coefficient, the middle line is the p-value for the correlation, and the bottom line is N. Values along the diagonal compare the seasons. The top line is the percent difference comparing warm season to cool season, the middle line is the p-value for the difference in means, and the bottom line is N.

[b] Median(IQR) concentration in $\mu g \cdot m^{-3}$

[c] Median(IQR) concentration in $ng \cdot m^{-3}$

[d] Median(IQR) concentration in $\#\cdot cm^{-3} \times 10^3$